Process development and process monitoring of landfill leachate treatment in combination with complementary long-term addition of process water from fermenter

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Abstract

Before transporting the landfill leachate to municipal wastewater treatment plant it has to be treated in a landfill leachate treatment plant, as it comprises high concentrations of ammonium. The elimination of ammonium load in the leachate is usually done by the combined processes of nitrification and denitrification with a specially adapted biocenosis in the activated sludge (AS). For each of the steps, specialized bacteria such as *Nitrosomonas*, *Nitrobacter* and *Paracoccus* are used to transfer the ammonia to gaseous nitrogen. The aim of this investigation was to find suitable process parameters for a complementary treatment of fermentation water from a biogas plant together with landfill leachate. The processed water of the biogas plant consists of a higher concentration of ammonium and carbon sources or easily degradable volatile fatty acids. It can save the usage of external carbon source (acetic acid) and additionally it could also compensate the missing volumes of leachate in times of low rain and low leachate flows. To maintain the high workload for the existing leachate treatment pilot plant (LTPP), a combined treatment of landfill leachate and process water is also of economic and of ecological interest. The long-term adaption process of the biocenosis needs to be done step-by-step. Innovative process monitoring is needed to prevent biocenosis collapse. In our study, we present our set-up, a closer look at the ongoing experiment and the long-term changes in the biocenosis.

1. Introduction

Since earlier days, landfills are most common methods of organized waste disposals and remained so in many regions of the world [1]. In Germany in Rheinische-Bergisch, Oberbergischer Kreis in Lindlar Remshagen, there is such a landfill, the so-called Leppe landfill. The Bergischer Abfallwirtschaftsverband (BAV) operates this landfill since 1982. The landfill site produces approximately 400 m^3 leachate water per day [2]. This leachate water has to be treated before transporting it to the municipal wastewater treatment plant in Bickenbach, as it contains a high amount of nitrogen compounds. A classical wastewater treatment plant alone cannot clean it, because those are mainly designed for the reduction of mainly carbon containing organic compounds.

In most cases, the main source of the landfill leachate water is rainfall, Ground-water inflow, surface water runoff, and biological decomposition also lead to the production of landfill leachate water [3]. The composition of the leachate can vary depending on the landfill age [4]. During the years, the landfill leachate changes itself due to the different decomposition in the plant (Table 1).

Table 1: Landfill leachate classification vs. age [5]

	Young	Medium	Old
Age (year)	<1	1-5	>5.0
pH	<6.5	6.5-7.5	>7.5
COD (g L^{-1})	>15	3.0-15	<3.0
BOD_5/COD	0.5-1	0.1-0.5	<0.1
TOC/COD	<0.3	0.3-0.5	>0.5
NH_3-N (mg L^{-1})	<400	400	>400
Heavy metals (mg L^{-1})	>2.0	<2.0	<2.0
Organic compound	80% VFA	5-30% VFA+ HA+FA	HA+FA

1.1 Activated sludge process

The activated sludge process is the most extensively used biological process for municipal and industrial wastewater treatments [6]. It is a process of utilizing microorganisms to convert organic matter into carbon dioxide, water, and inorganic compounds [7]. There are a lot of different bacteria and protozoa in activated sludge, the sum of all organisms and their life together in this ecological system is called “biocoenosis”. This biocoenosis describes the interaction of all organisms in the system. It is subjected to a continuous change in the amount and the composition of the bacteria as an adaption to varying substrates in their feed. Different process parameters can lead to a different composition. As an example, also the temperature dependency of bacteria could be mentioned. For different temperatures, different bacteria can survive and/or have their thermal optimum [8].

1.2 Nitrification and denitrification

Nitrification is the first essential step in the removal of the ammonium from landfill leachate water. The process in an aerated reactor with activated sludge in which ammonia is oxidized to nitrite (with ammonia-oxidizing bacteria and archaea) and nitrite to nitrate (nitrite oxidizing bacteria and archaea) is called nitrification. A simplistic chemical equation for the whole process is outlined in Table 2.

Equation 1: Nitrification [9]

Ammonia oxidation	$NH_4^+ + 1{,}5\ O_2 \rightarrow NO_2^- + H_2O + 2H^+$
Nitrite oxidation	$NO_2^- + 0{,}5\ O_2 \rightarrow NO_3^-$
Nitrification	$NH_4^+ + 2\ O_2 \rightarrow NO_3^- + H_2O + 2H^+$

Denitrification is the second step in the nitrogen elimination process. It is a microbial process of reducing NO_3 to Nitrogen gas (N_2) by facultative heterotrophic bacteria [6]. The denitrification process takes several steps with different enzymes. No oxygen but an external carbon source is needed. It can be expressed as a redox reaction.

Equation 2: Denitrification [9]

Denitrification $2\ NO_3^- + 10\ e^- + 12\ H^+ \rightarrow N_2 + 6\ H_2O$

With	NH_4^+	=	Ammonia	*with*	O_2	=	Molecular oxygen
	NO_2^-	=	Nitrite-Ion		H^+	=	Hydrogen proton
	NO_3^-	=	Nitrate-Ion		e^-	=	Electron
					H_2O	=	Water

1.3 Pilot plant for landfill leachate treatment

The pilot plant (semi-technical scale) used for landfill leachate treatment is a scale-down of the industrial plant from BAV and a scale-up of the lab scale leachate water treatment plant.

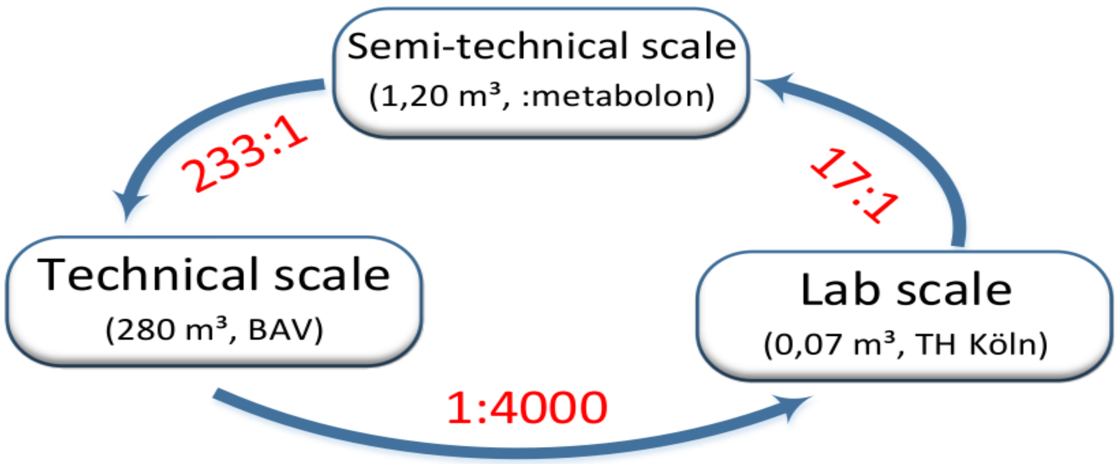

Figure 1: Different volumes of the plants and volumetric scale up factors [10]

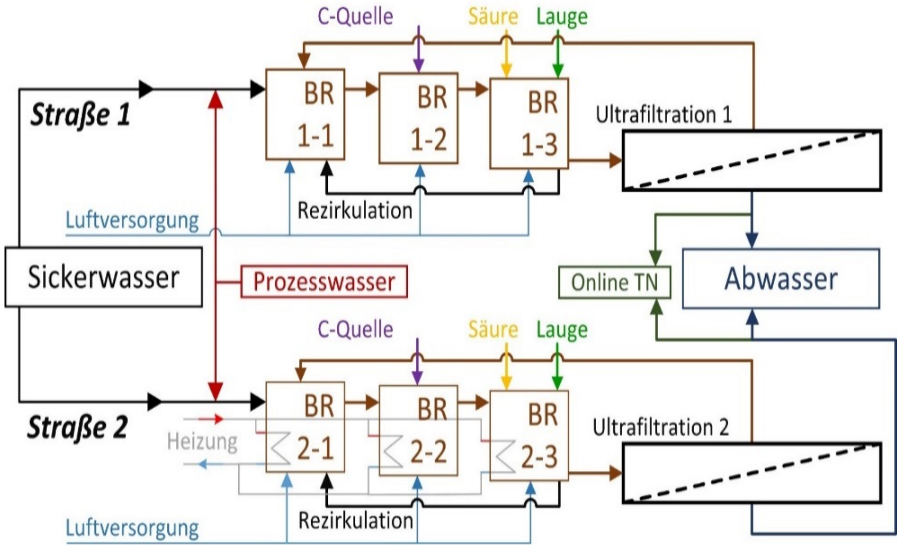

Figure 2: Schematic representation of Pilot plant [11].

As shown in Figure 2 the pilot plant for landfill leachate treatment was designed in two lanes (street 1 & street 2). A direct and valid scientific comparison between two different process strategies is possible. In the pilot plant data management software, automation with LabVIEW and enhanced analytical tools are used, as shown in following picture (Figure 3).

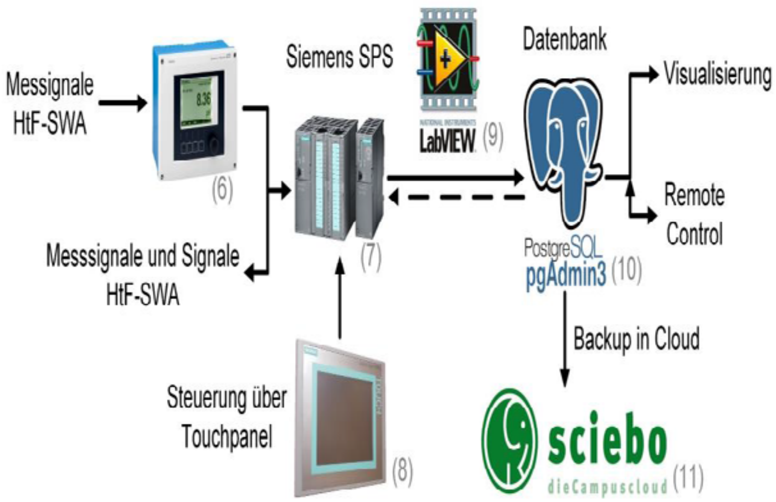

Figure 3: Simplified and general presentation of the data flow of the Pilot plant [11]

1.4 Process water from biogas plant

Generally, the water generated during the processes such as softening, demineralization biogas and others. operations are known as process water. According to the German law, purification of this contaminated water has to be done before transporting it to municipal wastewater treatment plant. In the area of wastewater purification, process water is defined as wastewater from sludge thickening and sludge dewatering on sewage treatment plants with anaerobic sludge stabilization [12]. In this study, the process water was obtained from the research site: metabolon i.e. from fermentation and composting plant (AVEA). The combined fermentation and composting plant went into operation in 1997 and utilized approx. 660,000 tons of bio-waste by the end of 2017. This resulted in a production of electricity. The completion of additional tunnel composting has increased the processing capacity from 45,000 tons to 75,000 tons since the end of 2018 [11].The anaerobic digestion is carried out according to so called Valorga method under mesophilic conditions (temperature about 40 ° C). The organic material is fermented with a residence time of about 28 days.

After some required separations, the obtained bio waste is transferred to a mixing unit. Subsequently, the stream is diluted with water and pumped into the fermentation tank. The cylindrical fermentation tank is divided by a middle wall with an inlet and outlet on opposite sides. The wall rotates around its own axis and pushes the biomass around in a circle until it overflows into a collecting container. The cylinder also consists of a plate with nozzles for blowing air from the bottom. As the biomass rots, it slowly moves upwards by the rotating wall but remains in the reactor for a certain period and the gas is expelled. The gas is collected at the top and the overflowing biomass is dewatered and processed. The gas is burned for energy. As shown in figure 4, the process water obtained after belt filtration was used in this study.

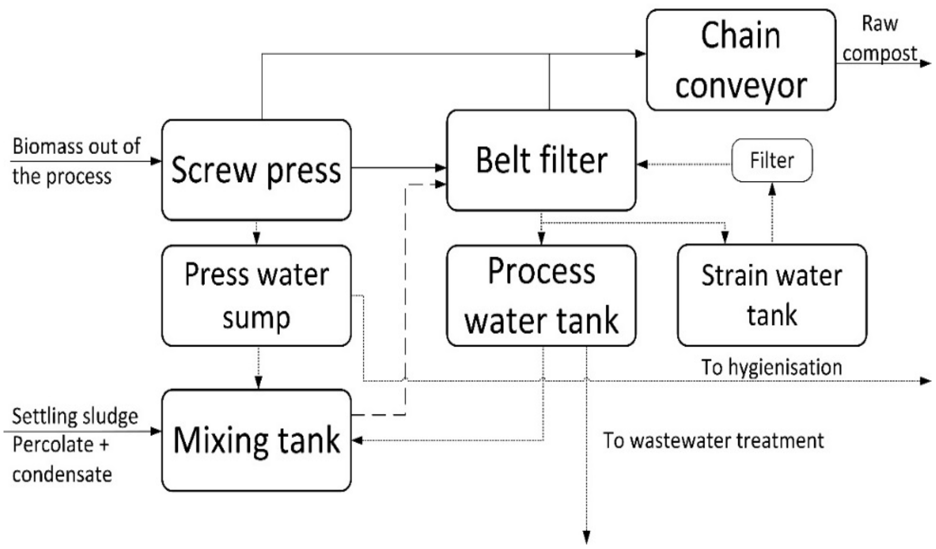

Figure 4: Process water tank was the sample collection point in biogas plant.

1.5 Objectives:

- Optimizing the leachate treatment pilot plant with two identical streets for the experiment of long-term adaptation of micro biocenosis with fermentation wastewater (process water).
- Monitoring the nitrogen metabolism in both streets during the entire experiment.
- Investigating the morphology of the activated sludge by light microscope.
- Examining the particle size of the activated sludge floc.

In this investigation street one (S1) was used to treat process water (PW) and leachate whereas, S2 (control) was used only for leachate treatment. The PW share was increased periodically in S1 from 1.01 % (v/v) to 48,78 % (v/v) (Phase 1) and then ammonium load was increased by increasing the inflow rate (stress test) (Phase 2). The NH_4-N load of 500 mg/L (approx) was maintained in both streets during phase 1. In phase 2 (stress test) NH_4-N load was gradually increased to examine the stress phase of biological process in pilot plant.

2. Material and Methods

During the leachate water treatment, several factors can influence the efficiency of the activated sludge process and can show a huge influence on the results. Certain parameters of the pilot plant, like pH, redox potential, dissolved oxygen are continuously monitored and maintained in the range for efficient treatment. Some of the necessary materials are mentioned in the following tables.

Table 2: Chemicals and equipment for pilot-scale tests

Chemical / unit	Manufacturer	Purity / Specs	item number
Acetic acid	Brenntag	60%, technically	-
Microbalance	Core	ACJ 320-4M	WB12AC0129
Microscope	Zeiss	Axio Lab A1	3136005267
Microscope (camera)	Zeiss	Axio cam 105 color	10000007517
Muffle furnace	Nabertherm	LT 9/11 / C450	352447
Particle analyser	Ankersmid	Eyetech	09501101
Phosphoric acid	Brenntag	75% strength,technically	-
Photometer laboratory	Hach & Lange	DR 6000	1524699
Photometer	Hach & Lange	DR 2800	1264930
Buffer solution	Merck	pH 7.00	1.99057.0500
Buffer solution	Merck	pH 4.00	1.09445.0500
Ultrapure water plant	Werner	Nanopure®	383337-113
Sulphuric acid	Merck	98%	1.12080.2510
Syringe Filters	MN	Chromafil CA_45 / 25	729027
Drying cabinet	Memmert	UN 55	B212.0472
RE water system	Werner	AQUADEM ®	20 SDF
Libra technical	PCE	TS-150	T150325024

centre			
Webcam	Vimtag	VT 361	1JFEGBP1JCA
Centrifuge laboratory	Hermle	Z 326 K	66130235
Centrifuge	Hettich	EBA 20, 6,000 rpm	008547
Landfill leachate		From technical scale plant	
Activated sludge		From technical scale plant	
Process water		Avea (fermentation and composting plant)	

Table 3: Analysis directly associated with the pilot plant

Device name	**Parameter**	**Manufacture**	**Installation**
Cerabar M PMC51	Level [%]	Endress + Hauser	BR 1-3, BR 2-3
Cerabar T. PMP 131	Pressure sensor [bar]	Endress + Hauser	UF S1 + S2
CondumaxCLS21D	Conductivity [µS / cm]	Endress + Hauser	Inlet S1 + S2 Sequence S1 + S2
Dipsy's CPA-4-0A	Redox potential [mV]	Endress + Hauser	BR 1-1, BR 2-1
ISEmax CAS 40D	NH_4+ , NO_3 , K^+ , Cl^- [mg / L]; pH	Endress + Hauser	BR 1-2, 1-3 BR 2-2, 2-3
Liquicap T FMI21	Level [%]	Endress + Hauser	B-2000
Liquipoint FTW32	Measuring leakage via conductivity	Endress + Hauser	10 m^3 tanks
Orbsint CPS11D	pH	Endress + Hauser	Inlet S1 + S2 Sequence S1 + S2
OxyMax COS61D	dO2 [mg / L]	Endress + Hauser	BR 1-1, 1-3 BR 2-1, 2-3
Promag 50H	Flow rate [L / min]	Endress + Hauser	Recirculation S1 + S2 Inlet S1 + S2
TMR 31	Temperature [° C]	Endress + Hauser	BR 1-1, 1-2, 1-3 BR 2-1, 2-2, 2-3
Turbimax CUS52D	Turbidity [FTU]	Endress + Hauser	Sequence S1 + S2
QuickTOCN ®	TOC, DOC	LAR	Offline
Multi meter	Various	WTW	handheld instrument

FDO925	DO_2 measurement	WTW	handheld instrument
SenTix® 940-3	pH measurement	WTW	handheld instrument

Table 4: Devices of the pilot plant

Device name	Parameter	Manufacture	Installation
Ecoline	ISM1079	Ismatec	Pump PW
Frequency converter for stirrers	-	Lenze ACTech	Tank PW
Gala 1000 PVT200	0-12.33 mL / h	Prominent	C source S1 + S2 Acid + alkali S1 + S2
MA II 5-230	Max: 95 L / min	Lutz	Logistics DSW / VAB
Movitec VSF6 / 5B	104.33 L / min	KSB	ultrafiltration S1 + S2
OXYFLEX-MT300 coarse-bubble	0.07 m² gassing area	Subratec	Membrane aerators of all reactors
Rapid R3	1,568 L / min	Verderflex	Logistics DSW
Stirrer	-	SIMIX	Tank VAB
SK 1282AFSH-63L / 4 GMF 0.18 / 78	Rotary-speed stirrer 100 ± 1 * 1	RVT	Agitator of all reactors Motor of all reactors
Smart S10	0-1.7 L / min	Verderflex	Inlet S1 + S2 recirculation S1 + S2
PLC * 2	S7-300	Siemens	HtF -SWA
PLC * 2	S7-300	Siemens	UF
Tank VAB	250 liters	-	Tank VAB
Touch panel	KTP 1000	Siemens	HtF -SWA
Touch panel	TP 700	Siemens	UF
Transport tanks (IBC)	1000 liters	Various	Logistics DSW
Ultrafiltration membranes	0.51 m^2	Memos	Ultrafiltration S1 + S2

2.1 Photometric tests

For the analysis of NH_4-N, NO_2-N, NO_3-N, and COD during biological treatment processes a photometric test is performed by a photometer with Hach-Lange cuvette test (see Table 5). 10 ml sample was collected in centrifuge tubes from the reactors twice a week and centrifuged for 5 min at 60000 Us^{-1}. The supernatant obtained after centrifugation were collected in plastic tubes for immediate analysis. If the concentration of the compounds (NH_4-N, NO_2-N, NO_3-N) is higher than the measurement range of the cuvettes (see Table 5), the samples were further diluted (1:2 or 1:5) with deionized water and then measured. Only 2-5 ml of the sample was utilized for the measurements. The remaining sample was stored in a freezer at -20 °C for further use and other analytics.

Table 5: Cuvettes used during the experiments from the company Hach

Measured variable (Ion)	Abbreviation	Method	Article No	Measuring range
Ammonium nitrogen (NH_4 -N)	NH_4-N	ISO 7150-1, DIN 38406 E5-1, UNI 11669: 2017	LCK 304 LCK 303	0.015-2.0 mg / L 2.0-47.0 mg / L
Nitrate Nitrogen (NO_3 -N)	NO_3 -N	ISO 7890-1-2-1986, DIN 38405 D9-2	LCK 340	5.0-35.0 mg / L
Nitrite-Nitrogen (NO_2-N)	NO_2-N	EN ISO 26777, DIN 38405 D10	LCK 342	0.6-6.0 mg / L
Total Phosphorus Phosphorus (PO_4-P)	PO_4 -P	ISO 6878-1-1986, DIN 38405 D11-4	LCK 348	0.5-5.0 mg / L
Chemical oxygen demand (COD)	CSB	ISO 15705	LCI 400	0.0-1000.0 mg / L

2.2 Dynamic Image Analysis by EyeTech

To have a better knowledge on activated sludge flocs in the pilot scale plant, the morphology of activated sludge flocs were examined by a dynamic image analyzer (EyeTech) from the company Ankersmid (The Netherlands). With this instrument, even floc size distribution can be evaluated. The floc size distribution gives required information to estimate the particle occurrence frequency in different size ranges. The sludge samples (3 mL) to be analyzed were diluted in 500 mL tap water and measured with ACM 104L fiber measurement cell by pumping the sample continuously through the cell during measurements. The activated sludge sample is collected from all six reactors in S-1 (PW Street) and S-2 (Control Street). The sludge samples were collected with a micropipette. To avoid shear stress to the sludge particles during sample collection, a small portion of the micropipette tip was removed using cutter. The video measurement mode was used in this work; there is a possibility for a user to set up a precise dynamic image analysis so that a method can be developed for measurements.

In this study, the following method was used for all measurements.

- Stirrer speed: Normal
- Dilution:1.5 ml sample in 500 ml tap water
- Illumination: Exposure: 2, Intensity: 8
- Image control: Gain:127, Contrast:255, Brightness:153
- Threshold Range: 0 – 100
- Remove Unfocused: Used, Focus Level = 8
- Pre-processing commands: contrast enhancement, high path medium, histogram equalization
- Morphological commands: remove touching frame objects, fill holes

2.3 Morphological investigation of activated sludge by light microscope

From the first reactors of each street of the pilot plant 50 mL of activated sludge were collected. With the help of a pipette few drops of sludge was placed on a glass slide and a cover slip was placed on the drops; it was ensured that there were no bubbles trapped. The slide was then placed under the Zeiss light microscope (see table 2) for examination. A camera (Axio cam 105 color) from Zeiss was used to capture the images of microorganisms in activated sludge. Throughout the study 40x magnification was used to visualize the microorganisms.

3. Results and discussion:

The results of the NH_4-N concentration in pilot plant over the entire test duration of 532 days are shown in following graph.

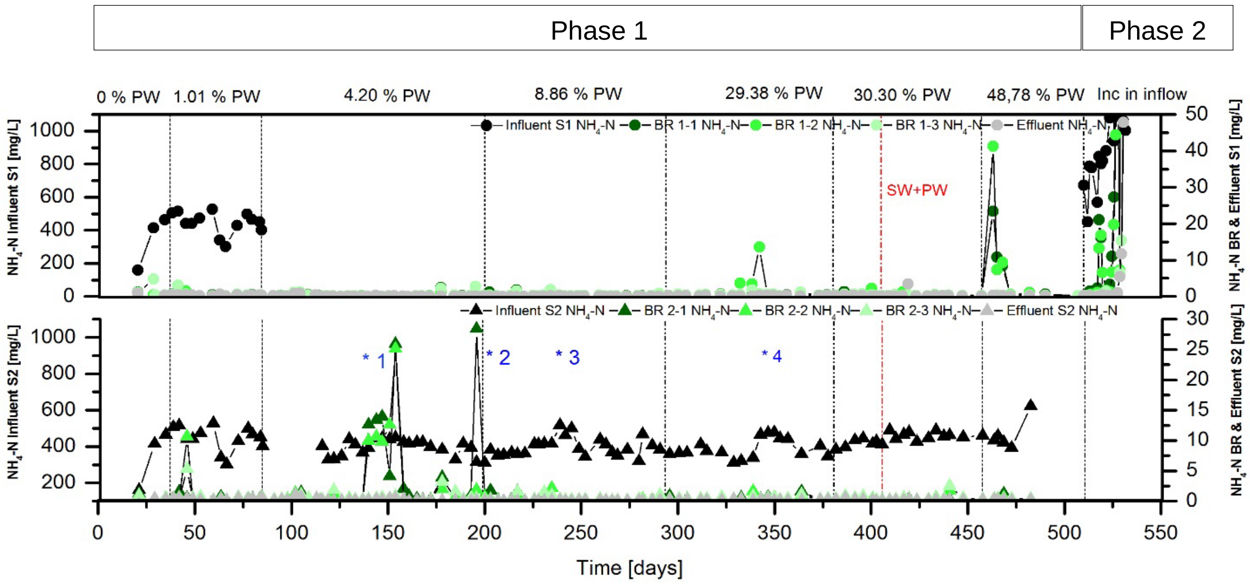

Figure 5: Overview of NH_4-N concentrations over the entire experiment.

The disturbances shown in Figure 5 are explained as follows:

- * 1: Power failure: Failure of various parts of the system during automatic restart.
- * 2: Recirculation pump malfunction: recirculation was stopped.
- * 3: Malfunction of the pump of the PW: Increased volume flow in the Pilot plant.
- * 4: Failure of the compressor for air in the PW Street.

Apart from these system disruptions, a stable operation of the pilot plant was achieved.

As shown in the Figure 5, the complete experiment was divided into two phases. During phase-1 NH_4-N, concentration was approximately 500 mg/L in both street, In PW street the volume of leachate was reduced and PW was used instead to maintain the same ammonium load in both streets. Periodically the volume of leachates were decreased and volume of PW was increased. As shown in Figure 5, PW concentration was increased from 1.01 % (v/v) to 48.78 % (v/v). Later on, the NH_4-N load was increased in street one by increasing the volume of inflow. From day 32 to 407, required volume of PW and leachate were pumped with two different pumps. Later in order to avoid the malfunctioning of PW pump, from day 408 both leachate and PW were mixed in 250 Liters tanks and then pumped accordingly. Except in few cases of disturbances as shown above, NH_4-N degradation was almost comparable in both streets. There was no NH_4-N accumulation in the bioreactors. The micro biocenosis of the activated sludge was successfully adapted by successively increasing the proportion of PW at a constant nitrogen load. The plant could be operated stably up to a share of 48.78 % PW in the total volume flow for 25 days. After a necessary change of the streets by conversion measures and restarting of the plant, the performance of the micro biocenosis was tested in a stress phase. The volume flow was increased gradually with a constant proportion of PW. In the last step of the increase, the inflow of the plant was three times to that of the regular experiment. Furthermore, it was found during the stress test that the Pilot plant functioned stably up to 1.6 times increase in feed stream.

On the other hand, the results obtained from the EyeTech (Figure 6) showed the particles with higher average ferret diameter (AFD) in S1 and comparatively lower AFD particles in S2. The results shown above are the average of 23 measurements of each bioreactor respectively. The bigger particle size in S1 might be due to the large sized waste particles in PW as well as due to green waste. AFD of the sludge particles in S1 were in between 20-520 µm (approximately) whereas, the particles in S2 were in between 20-350µm (approximately). The presence of particles with higher AFD did not showed any negative influence on the nitrogen metabolism.

The microscopic images shown above in the Figure 7 were taken during a period of 8 months. These results showed more filamentous bacteria in S2 than S1. The presence of PW might be toxic to the filamentous bacteria. It is recommended to do further research in these criteria to find out the precise reasons behind the influence of PW on the filamentous bacteria present in the activated sludge of the technical scale plant.

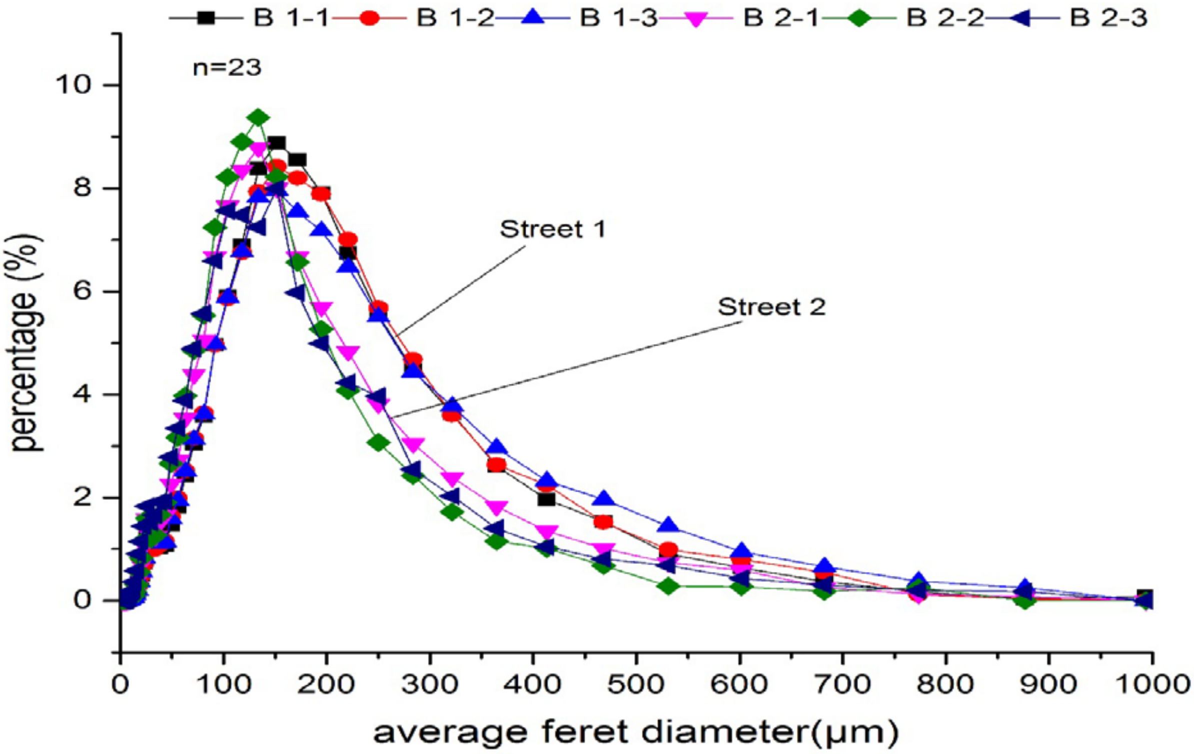

Figure 6: Particle size of the activated sludge flocs in S1 & S2 of pilot plant.

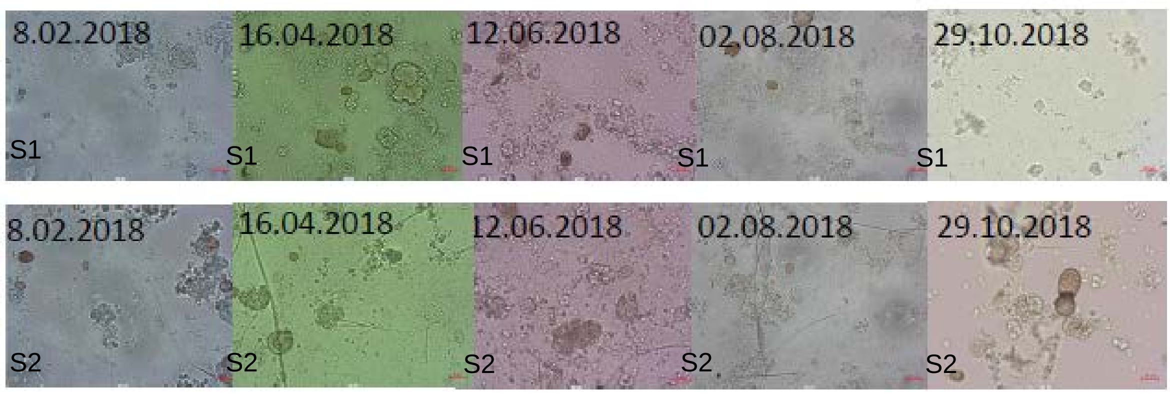

Figure 7: Microscopic images of the activated sludge in the nitrification reactors.

Conclusions

The micro biocenosis of the activated sludge was successfully adapted by successively increasing the proportion of PW at a constant nitrogen load. There were no differences in the process stability of the street with PW compared to the control street. The plant could be operated stably up to a share of 48.78% PW in the total volume flow for 25 days. After a necessary change of the Street and restarting the pilot plant, the performance of the micro biocenosis was tested in stress phase. The volume flow was increased progressively with a constant proportion of PW. In the last step of the increase, the inflow of the plant was three times to that of the regular experiment. Furthermore, it was found during the stress test that the pilot plant functioned stable. The stable plant performance can be confirmed by measurements of the relevant groups of organisms by CenoTox measurements [13].

Although the method still needs to be optimized in terms of aeration time, the analytical process used to assess process stability is helpful. The detailed results regarding this experiment can be found in [13]. The results of EyeTech showed higher AFD particles in S1 but they did not have a negative influence on the ammonia degradation. The above results showed that the combined treatment of PW from fermentation reactor of biogas plant with leachate is possible by suitable adaptation of the biocenosis.

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